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Final Technical Report (May 1, 1996 through December 31, 1998)

"Electron Excitation of Atoms and the Intermediate Stretched Atom States" AFOSR Grant F49620-96-1-0137

The research conducted under this project were directed toward electron collisions with atoms with emphasis in understanding the roles of the intermediate stretched atom states in the excitation processes.

- To experimentally determine electron-impact excitation cross sections with the optical 1. method, it is necessary to measure all transitions out of a level (the apparent cross sections), as well as the cascades into the level. In the case of the ten levels within the 3p⁵4p configuration of argon, the emissions into the lower levels (3p⁴4s) lie in the visible and near infrared so that we can measure the apparent excitation cross sections for these levels by a conventional monochromator/photomultiplier system. The cascade is due to emissions into the 3p54p levels from such highly excited states as the levels of 3p53d and 3p⁵5s configurations which are in the infrared, hence these cascade cross sections have not been measured previously. However, with our Fourier transform spectrometer we are now able to measure the infrared cascade emission cross sections. Such measurements are important for the following reasons. Firstly, these infrared cascades are produced by emissions from the highly excited states of the 3p⁵ns and 3p⁵nd configurations, thus acquisition of the infrared cascade cross sections allows us to probe into these highly excited states. Secondly the cascade emission of cross sections enable us to perform a complete cascade analysis and obtain the direct excitation cross sections for all ten levels of 3p⁵4p from the apparent excitation cross section data. The percentage cascade contribution to the apparent excitation cross section varies greatly from one level to another. Our results indicate that accurate measurements of the cascade are essential to obtain reliable direct excitation cross sections from optical studies.
- In an usual electron-beam excitation experiment conducted at low gas pressure (a few 2. mTorr), the intensity of the radiation emitted by the excited atoms is proportional to the gas pressure and the measured apparent excitation cross section, which is proportional to the measured intensity divided by the gas pressure, is independent of the gas pressure as it should be. At higher pressure secondary processes begin to set in so that the measured apparent excitation cross section depends on (usually increases with) the gas pressure. Such a pressure effect is often an indication of secondary processes. For argon this pressure effect has been observed at pressures as low as 1 mTorr or less, but the origin of this pressure effect was not known. In our experiments we found both the apparent excitation cross sections and the cascade cross sections to be pressure-dependent. However upon subtracting the cascade terms from the apparent excitation cross sections, the resulting direct excitation cross sections are independent of the gas pressure in the range of 0.5 to 4 mTorr. Therefore the observed pressure dependence of the apparent excitation cross sections of the 3p⁵4p levels is due to pressure-dependent cascade into these levels but not collisional excitation transfer into these levels.

- We have conducted research toward understanding the origin of the pressure dependence 3. described in the preceding paragraph. We determine that this pressure dependence can be traced to radiation trapping in the following manner. Consider an excited level-a with J=1 that decays in an optically allowed transition not only to the ground level g (J=0) but also to a number of other levels b, c, ... with J=0 or 2. Reabsorption of the a→g resonant transition by a ground-level atom with the subsequent decay into level b results in a pressure-dependent effective branching ratio for the a-b transition and therefore a pressure dependent a-b emission cross section. Optical cross sections from such resonant (or optically allowed) level-a are known to exhibit pressure dependence. However, levels not optically connected to the ground level, such as the 3p54p levels of argon, can also display pressure dependence due to cascade from higher levels. For instance although a 3p⁵4p level with J=0,1, or 2 is not optically coupled to the ground level (same parity), it may receive significant cascade from the J=1 levels of the 3p⁵4s and 3p⁵3d configurations which do decay to the ground level, so that the total population of this 3p⁵4p level is affected by reabsorption and its apparent excitation cross sections display a pressure dependence.
- 4. To further pursue the issue of radiation trapping and its effects on excitation cross sections, we have measured the optical cross sections of the 5¹P→2¹S transition of He at 100 eV at various pressures from 0.05 to 50 mTorr. We are able to explain quantitatively the observed pressure dependence very accurately using the radiation trapping model described in the preceding paragraph.
- 5. The use of Fourier transform spectroscopy to study the highly excited levels and stretched atoms has been extended beyond the experiments described in items 1 and 2 above. We have developed special techniques to measure cross sections for emissions from the highly excited levels of the 3p⁵5p, 3p⁵6p, 3p⁵7p, and 3p⁵4f, 3p⁵5f configurations into the 3p⁵3d and 3p⁵5s levels.
- The nF states (orbital angular momentum ℓ=3) of the helium atom play a unique role in 6. electron-impact excitation. The nF levels are nearly degenerate with the D levels. Even when the incident electron is "far away" from the atom, the Coulomb interaction between the incident electron and the target atom may be sufficient to mix the F and D states, corresponding to a stretched atom. Thus the excitation cross sections of the F levels may be influenced by the cross sections of the D levels and differ from what may be expected of just the "unstretched" F levels. Another special feature is that the energy spacing between an nF level and the n¹P level of the same n is much smaller than the thermal energy, e.g., 40 cm⁻¹ for n=4, 21 cm⁻¹ for n=5, and 12 cm⁻¹ for n=6. An atom in the n¹P level (populated by electron excitation) may easily transfer into the nF level upon colliding with a ground-state helium atom. Thus in an electron-beam excitation experiment the nF levels may be significantly populated by collisional transfer from the n¹P levels in addition to a one-step electron-impact excitation from the ground level. Indeed, the excitation transfer (from the n¹P levels) is believed to be the major mechanism for populating the nF levels. The LS-coupling applies to the S,P,D levels of

helium, but not to the F levels because for the F levels the exchange interaction between the 1s and nf electrons is much smaller than the spin-orbit coupling of the nf electron. In other words the F-state wave functions are represented by mixtures of singlet and triplet eigenfunctions. Because of this singlet-triplet mixing, the nF levels can cascade to both the ¹D and ³D levels. In this manner the n¹P-nF transfer propagates down to the ¹D and ³D levels. One manifestation is the observed change in the shape of the excitation functions of the ¹D and ³D states as pressure is increased above 20 mTorr.

In spite of its importance in atomic collisions, very little had been done in way of measurements of the cross sections for electron-impact excitation into the nF levels. The main reason is that in order to detect the atoms excited into the nF levels, one has to measure the radiations emitted by the nF atoms which are in the infrared, outside the spectral region covered by the conventionally used photomultiplier tubes. Since we have recently equipped our laboratory with the capabilities of Fourier Transform spectroscopy, we are in a position to measure the infrared radiations emitted by the nF atoms produced in an electron-beam experiment. In this manner we have measured electron-impact optical cross sections for the 4F→3D, 5F→3D, and 6F→3D emissions at various gas pressures form 3 to 50 mTorr and incident electron energies from threshold to 200 eV. The measured emission cross sections show very strong pressure dependence in agreement with the transfer mechanism from the n¹P levels. Analysis of this pressure dependence in agreement with the transfer mechanism from the n¹P levels. Analysis of this pressure dependence enables us to determine, for the first time, the electron-impact excitation cross sections into the 4F, 5F, and 6F levels. We have also obtained the $n^{1}P \rightarrow nF$ collisional excitation transfer cross sections for n=4, 5, 6, 7.

7. Publications

- (a) "Measurement of Electron-Impact Excitation into the 3p⁵4p levels of argon using Fourier-Transform Spectroscopy" by J.E. Chilton, J.B. Boffard, R.S. Schappe, and C.C. Lin, Physical Review A <u>57</u>, 267 (1998).
- (b) "Electron-Impact Excitation and Collisional Transfer into the nF Levels of Helium" by J.E. Chilton and C.C. Lin, Physical Review A <u>58</u>, 4572 (1998).
- (c) "Measurement of Electron-Impact Excitation into the 3p⁵3d and 3p⁵5s Levels of Argon using Fourier-Transform Spectroscopy" by J.E. Chilton and C.C. Lin, Physical Review A, accepted for publication.